



Mechanically robust, stretchable organic solar cells via buckle-on-elastomer strategy

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ABSTRACT

In recent years, the exploitation of stretchable organic solar cell (OSC) has attracted significant research interests due to the rapid progress of wearable electronics. However, the development of a stretchable OSC is quite challenging since it has a strict requirement for the mechanical deformability and durability of each constituent layer in device. In this work, we successfully fabricated an efficient, stretchable inverted OSC by adopting a buckle-on-elastomer strategy, for which an ultrathin poly(ethylene naphthalate) (PEN) substrate coupled with a pre-strained (100%) 3M elastomeric tape was employed as the device substrate. Owing to the pre-strained status of the elastomer, the ensemble wrinkle will be formed in response to accommodate the strain once the pre-strain was released, which can afford the derived OSC with a much improved mechanically robustness and stretchability. As a result, we demonstrated that a pristine efficient (PCE: 5.61%) OSC using such buckling scaffold can remain its 74% efficiency under 30% compression and, more importantly, can still retain its 64.3% efficiency after 50-cycle compression-stretching testing from 0% to 30% compression. Besides, the effects of mechanical deformation and durability on the electrical performance are also investigated. This work proves that the buckle-on-elastomer strategy can be a good solution for realizing efficient OSCs with reasonably good mechanical durability, revealing great potential serving as an ultrathin and lightweight power source for wearable device applications.

1. Introduction

Due to the rapid progress of wearable device applications, the demand of portable power supply system was tremendously increased in recent years. To develop a power supply system compatible to the wearable electronic device has become an urgent task nowadays [1–4]. In response to this, the exploitation of stretchable organic solar cell (OSC) has emerged from various existing photovoltaic techniques since it possesses several advantages such as solution printable capability, well flexibility, and high power-per-weight property [5–8].

In general, the device configuration of a typical OSC is composed of a bottom transparent electrode, charge-transporting interlayers, photo-active layer, and a top electrode [9,10]. To develop an intrinsically stretchable OSCs, each constituent layer in the device must have certain ductility [11]; meanwhile, the contact and the electronic properties at the associated interfaces in the device need to be considered as well [12]. All these factors impose a formidable challenging for the development of stretchable OSCs. For example, the polymer-fullerene bulk-heterojunction systems generally possess poor ductility (break at an elongation of 0.30%) [7], which restrains the stretchability of the

derived OSCs. In this regard, in the earlier stage, the researchers introduced a polymer donor with higher elasticity to address this deficiency [13–15]. For instance, Savagatrup et al. have utilized poly(3-heptylthiophene) (P3HpT) with a longer alkyl side-chain to endow the derived BHJ (P3HpT:PCBM) with an improved ductility than the parent P3HT:PCBM BHJ, for which the P3HpT:PCBM blend possessed a maximum tensile modulus of 0.07 ± 0.01 GPa whereas the P3HT:PCBM blend owned a larger value of 1.09 ± 0.15 GPa [8].

Besides the engineering of polymer donors, replacing the small molecule acceptor, PCBM, with a polymer acceptor seems to play a profounder role in improving the ductility of the derived BHJ since the easy aggregation of PCBM will hugely deteriorate the BHJ morphology during the compression-stretching cycle, leading to poor stretchability and resulting performance. Kim et al. have recently successfully demonstrated a high-performance stretchable OSC (PCE: 6.64%) using an all-polymer BHJ layer (PBDTTTPD:P(NDI2HD-T) [7], representing an important step for stretchable OSCs from the material level. They revealed that the all-polymer BHJ film owned a well ductile nature, exhibiting no crack until a high elongation of 7% and withstanding 150-bending-cycle with a bending radius of 1.5 mm.

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On the other hand, it is also essential to develop stretchable electrodes and device scaffold [16]. However, regarding the challenging in developing intrinsically stretchable electrodes, several methods from the device level were exploited to indirectly develop stretchable OSCs to date [17–19]. Among which, mechanical buckling scaffold is most widely adopted method for constituting stretchable OSCs since the preformed buckles can mediate the stretching and compressive strain [20–24]. Such method has also been utilized to develop other mechanically robust and stretchable electronics, such as sensors, field-effect transistors (FETs), and polymer light emitting diodes (PLEDs) [17,18,25,26]. In general, a pre-strained elastomer was employed as the substrate and the completed device stack fabricated on an ultra-thin ($< 10 \mu\text{m}$) polymeric substrate was then adhered onto the pre-strained elastomer to form random buckling [27–30]. The most important requirement of this method is that the fabricated electronic device must be highly flexible to sustain the mechanical deformation during the compression-stretching testing. Lipomi et al. first demonstrated that the stretchable P3HT:PCBM BHJ OSC using pre-stretched PDMS showed an PCE of 2.0% under the strain of 22.2% [31]. Soon after, Kaltenbrunner et al. have further modified the fabrication process by using this buckling scaffold showed improved performance of 4% under more than 20 compress-stretching cycles from 0% (original size) to 50% quasi-linear compression [27]. In addition, O'connor et al. fabricated devices that retained at least 80% of their initial efficiency up to 1000 cycles on the skin and in outdoor sunlight, showing good mechanical stability [32].

From the abovementioned achievements, it shows the success exploitation of efficient stretchable OSCs necessitates the integration of material and device engineering. In this study, we have utilized the buckle-on-elastomer strategy to prepare mechanically robust, stretchable inverted OSCs with much improved efficiencies by using more advanced BHJ system. The completed flexible device stacks were first fabricated on a PEN substrate ($2.5 \mu\text{m}$ thick) and then were attached onto a pre-strained 3M elastomeric tape as illustrated in Fig. 1. As

shown, once the pre-strain force applied onto the elastomer is released, random buckling scaffold is formed in response to accommodate the strain. As a result, we demonstrated that a pristine efficient (PCE: 5.61%) OSC using such buckling scaffold can still preserve a high PCE of 4.13% even under 30% compression. The effects of mechanical deformation and durability on the electrical performance are investigated to reveal that such good mechanically robustness stems from the intact film morphology under compression. More intriguingly, we demonstrated that such device can still retain its 64.3% efficiency after 50-cycle compression-stretching testing from 0% to 30% compression. This result disclosed that the stretchable inverted OSCs using random buckling architecture can have an outstanding compression-stretching endurance, revealing promising potential for applications in large-scale and wearable OSC production.

2. Experimental section

2.1. Materials

Polyethylene naphthalate (PEN) foil (Teonex[®] Q71, $2.5 \mu\text{m}$ -thick) was used as the substrate and 3M VHB 4905 was used as the elastomeric tape. A layer of 500-nm-thick indium tin oxide (ITO) was sputtered onto PEN by Tze Chiang Foundation of Science and Technology. Zinc acetylacetonate hydrate ($\text{Zn}(\text{C}_5\text{H}_7\text{O}_2)_2 \cdot x\text{H}_2\text{O}$), methanol (anhydrous, 99.8%), ethanol (absolute, 99.8%), [6,6]-Phenyl C_{71} butyric acid methyl ester (PC₇₁BM, 99.5%), 1,2-dichlorobenzene (anhydrous, 99.8%), 1,8-diiodooctane (DIO, 97%), and molybdenum(VI) oxide (MoO_3) were purchased from Sigma-Aldrich. Poly [(9,9-bis(3-(N,N-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)] (PFN) and poly [4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b;4,5-b']dithiophene-2,6-diyl-alt-(4-(2-ethylhexyl)-3-fluorothieno[3,4-b]thiophene-)-2-carboxylate-2,6-diyl] (PTB7-Th) were purchased from Luminescence Technology Corp (Taiwan).

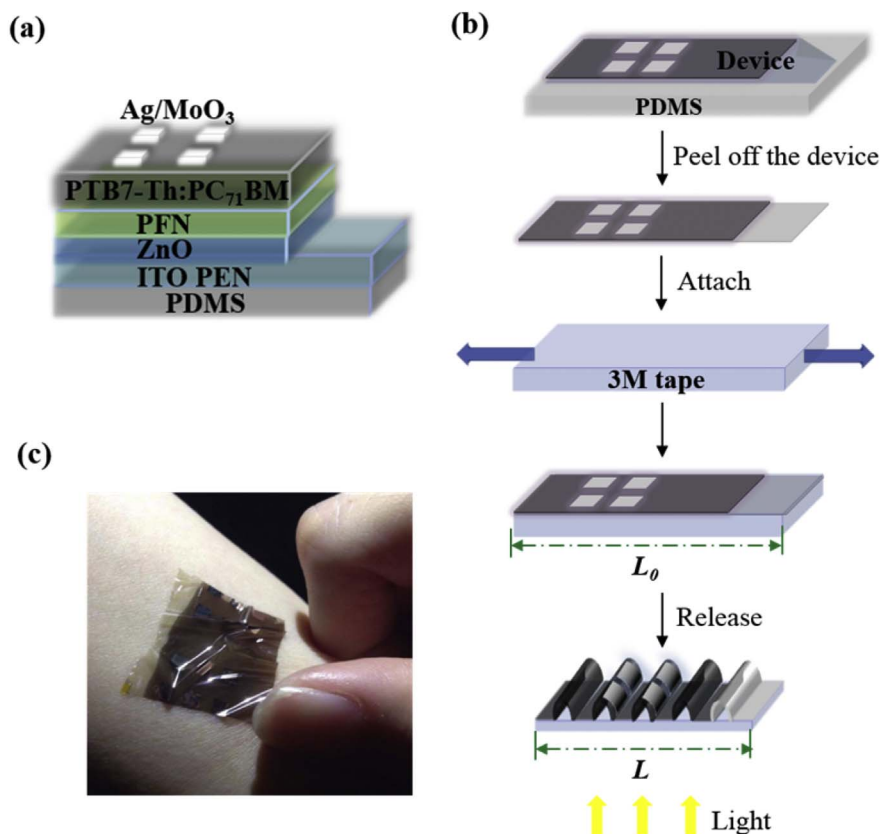


Fig. 1. (a) Device configuration of the fabricated solar cell device. (b) Illustration of the fabrication procedures of the studied stretchable solar cell device in this work, wherein a pre-strained (100%) 3M elastomeric tape was used and the prepared device stack was attached on top of it. Sequentially, the relaxation of the pre-strained elastomer developed the buckles. (c) A real photo portrayed a fabricated stretchable device attached onto the skin.

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